# Atomically Precise GaAs/AlGaAs Quantum Dots Fabricated by Twofold Cleaved Edge Overgrowth

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The formation of a $7 \times 7 \times 7\ \mathrm{nm}^3$ size GaAs quantum dot (QD) at the intersection of three quantum wells is demonstrated for the first time. Intense radiative recombination between zero-dimensional states in the QDs is clearly identified by microscopic photoluminescence ($\mu$PL). In contrast to the inhomogeneously broadened quantum well and quantum wire signals originating from the complex twofold cleaved edge overgrowth structure, the strongly spatially localized QD response is characterized by spectrally sharp lines in $\mu$PL excitation spectra with a linewidth below 70 $\mu$eV. [S0031-9007(97)04043-X]

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Semiconductor quantum wires (QWRs) and dots (QDs) with quantum confinement of charge carriers to one (1D) or zero dimensions (0D), respectively, have attracted extensive research activities recently, as they possess unique linear and nonlinear optical properties compared to systems of higher dimensionality. While several growth methods are well suited for the preparation of two-dimensional (2D) quantum well (QW) layers, the introduction of additional confinement by lateral patterning severely affects the optical quality of the structures and typical photoluminescence (PL) linewidths of single QDs exceed 0.5 meV [1–3]. In addition, substantial inhomogeneities in composition and size within single or arrays of low-dimensional structures cannot be avoided. Extremely narrow PL linewidths of less than 50 $\mu$eV, comparable to those of atomic spectra, have been observed in natural QDs which form due to well width fluctuations in narrow QWs [4–7] as well as in self-assembled QDs which are caused by a thermodynamically driven instability of the planar layer growth mode in strained systems [8–11]. As a result of the size and shape distribution present in such QD samples, however, emission from ensembles of QDs is dominated by inhomogeneous broadening and obscures the properties of the individual objects. Cleaved edge overgrowth (CEO), a molecular beam epitaxy (MBE) technique that uses high-quality regrowth on the cleaved edge of a multilayer sample [12] has proven to be a powerful technique for the fabrication of T-shaped QWRs which form at the intersection of two QWs [13]. The high perfection of these structures led to the demonstration of enhancement of exciton binding [14,15] and lasing from excitons in 1D [14]. As predicted in Ref. [16] and theoretically modeled recently [17], CEO can also be employed to produce QDs, in which charge carriers are confined in all three dimensions due to atomically precise QW potentials, and that are, therefore, highly uniform in size and shape.

In this Letter we describe the optical properties of a linear array of 22 almost identical GaAs QDs which have been produced by twofold CEO for the first time. The existence of 0D quantum mechanical bound states at the right angle intersection of three QWs is visualized in Fig. 1. Similar to the formation of a 1D state at the T-shaped intersection of two QWs [18] "expansion" of the electron and hole wave functions into the third QW due to confinement relaxation leads to a lowering of the corresponding ground state energy. Carriers which can move freely in two directions within the QWs and along one direction in the QWRs are completely localized in such QDs. If the layer sequence grown along the first growth direction consists of multiple QWs, the same number of QDs, connected via a single QWR results. Optical emission from

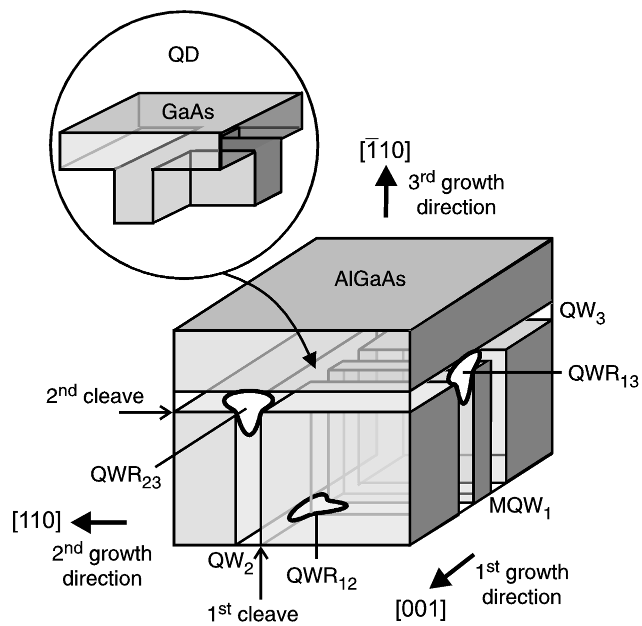


FIG. 1. Schematic illustration of the quantum dot structure (not to scale) obtained after three growth steps separated by two *in situ* cleaves. The junction of three quantum wells and wires, at which a quantum dot forms, is shown in the magnified part of the figure. The T-shaped contours are lines of constant probability for electrons confined in the quantum wires.

  

such QDs and the different QWRs and QWs is presented, clearly resolved both spatially as well as spectrally, and allows for the first time a direct comparison of all types of low-dimensional structures in one sample. The perfection with which these QDs are made is manifested by the occurrence of 70 $\mu$eV sharp lines caused by ground state excitonic transitions in 0D structures, not previously witnessed in QDs whose position and size is under detailed control through the fabrication process. Pronounced excitation of individual QDs under resonant pumping of QD states which are slightly above or below in energy is interpreted in terms of coupling between the 0D states through the connecting QWR. It thus appears that the twofold CEO QDs are ideally suited to perform fundamental studies on a novel class of well isolated or controllably coupled 0D objects.

The multiple QD structure consists of a 22-period $GaAs/Al_{0.35}Ga_{0.65}As$ multiple QW ($MQW_1$) structure with well and barrier thicknesses of 7 and 30 nm, respectively, which was 2 times *in situ* cleaved along the [110] and [$\bar{1}$10] cleavage planes and subsequently overgrown with two 7 nm wide GaAs single QWs ($QW_2$, $QW_3$) and $Al_{0.35}Ga_{0.65}As$ barriers. Details of the three step growth procedure are similar to those described in Ref. [14]. As can be seen in Fig. 1 three types of QWRs ($QWR_{12}$, $QWR_{13}$, $QWR_{23}$), resulting from intersections of $MQW_1$ with $QW_2$, $MQW_1$ with $QW_3$, and $QW_2$ with $QW_3$ form. At a length of about 800 nm 22 equally spaced and nominally identical QDs, each of which is constituted by the junction of three QWs, are located along $QWR_{23}$ about 930 nm below the (001) sample surface. The distances of the QD array from the (110) and ($\bar{1}$10) sample surfaces are approximately 340 and 140 nm, respectively. Low-temperature (5 K) microscopic photoluminescence ($\mu$PL) and $\mu$PL excitation ($\mu$PLE) spectroscopy is performed using a triple-grating Raman spectrometer equipped with a liquid-nitrogen cooled charge-coupled-device detector and a cold-finger He cryostat mounted on a $xyz$-translation stage. The positioning accuracy is about 50 nm. The sample is excited by a tunable dye laser whose beam is focused by a microscope objective (numerical aperture 0.75). PL light is collected by the same objective and passes a pinhole at an image plane which results in a submicron PL probe size.

A typical $\mu$PL spectrum recorded from the (001) sample surface in the vicinity of the QDs is displayed in Fig. 2. As a result of the large excitation area indicated by the circles in Fig. 2, which is unavoidable in diffraction limited spectroscopy, and exciton diffusion over a length of about 0.5 $\mu$m [19], the carriers, most of which are generated in the QW regions, recombine in the quantum dots, wires, and wells. Comparison with spectra taken at positions excluding excitation of the dots or wires (dashed circles) unambiguously identifies the PL response observed at 1589 meV and in the range of 1570–1580 meV as emission of $MQW_1$ and the two wells $QW_2/QW_3$. The PL

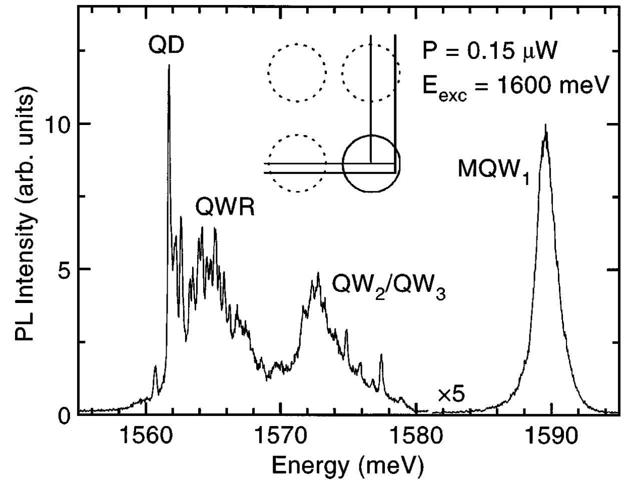


FIG. 2. $\mu$PL spectrum recorded from the (001) quantum dot sample surface (first growth surface) at a position indicated by the solid circle in the inset. The diameter of the circle corresponds to the spot size used in the experiment.

energies of $QW_2$ and $QW_3$ of 1574 and 1576 meV determined independently in this way almost coincide, but are, as a result of the large heavy hole mass for $\langle 110 \rangle$ type confinement directions, well below the $MQW_1$ response. Emission of the three types of QWRs present in the QD structure is detected around 1565 meV. Again, the PL maxima of $QWR_{12}$ and $QWR_{13}$, determined by moving the detection window away from the QD region, coincide within about 1 meV. With the high spatial resolution used for these experiments we find that the QWR PL and partly that of $QW_2$ and $QW_3$ starts to decompose into sharp lines as a result of exciton localization. In the QD region additional sharp lines appear at the low-energy tail of the QWR signal in the energy range of 1560–1563 meV, which we attribute to exciton recombination in the individual dots. This indicates that the QD states are about 4 meV deep with respect to the QWR states, a value which agrees well with the calculated 0D-1D exciton energy separation of 6 meV for a balanced structure [17], i.e., a structure where all QW ground transition energies exactly match and where therefore maximum 0D localization is obtained. It should be noted at this point that the binding energy of excitons to QDs fabricated by twofold CEO is not intrinsically that small. The use of narrower QWs and barriers made from pure AlAs should increase the depth of the 0D states to more than 20 meV as already demonstrated for T-shaped QWRs [15].

In order to identify these sharp peaks as originating from the QDs we have performed a detailed three-dimensional PL mapping of the sample. Figure 3 shows $\mu$PL spectra taken at different positions from the ($\bar{1}$10) sample surface together with grey scale intensity plots (pixel size 200 × 200 $nm^2$) at selected PL energies. For the position chosen in the upper panel of this figure, marked by a white cross in the grey scale image, emission from $MQW_1$ and $QWR_{13}$

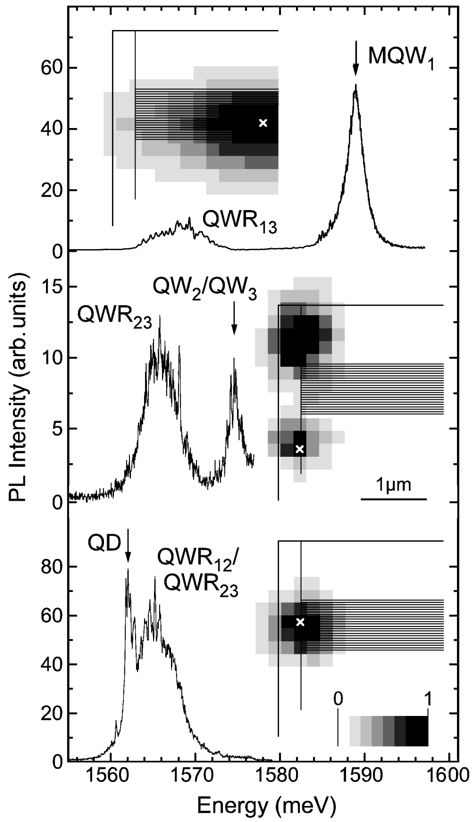


FIG. 3. $\mu$PL spectra ($E_{\mathrm{exc}} = 1600$ meV, $P = 0.15\ \mu$W) and grey scale intensity plots recorded from the ($\bar{1}10$) quantum dot sample surface (third growth surface). The positions on the sample where the spectra were taken are indicated by the white crosses in the grey scale images which are superimposed on a schematic view of the sample. The arrows define the spectral position of the corresponding grey scale plot.

can be detected. The grey scale intensity plot recorded with the detection system kept fixed at the MQW$_1$ PL line, indicated by an arrow in the spectrum, clearly reveals quenching of the multiple quantum well emission in the vicinity of the QDs due to exciton diffusion into wires and dots. The fact that the MQW$_1$ PL signal is not accordingly suppressed at positions away from the QDs but still on top of QWR$_{13}$ is a result of the finite excitation and collection depth of our probe which exceeds the exciton diffusion length. In accordance with the detection position used for the spectrum displayed in the middle panel of Fig. 3, only signals originating from recombination within the two single QWs (QW$_2$, QW$_3$) and the single wire (QWR$_{23}$) which is formed by these two wells can be detected. Again, bleaching of the QW emission in the vicinity of the multiple quantum wires and dots is observed, as can be seen in the corresponding grey scale plot. Exclusively at the intersection of the wells and wires, i.e., on top of the QD array, the characteristic sharp peaks, interpreted before as PL originating from the QDs, emerge (lower panel of Fig. 3). Although the relative intensities of these peaks have changed with respect to the spectrum shown in Fig. 2, their energetic positions exactly coincide. The intensity decrease of the QD emission is in all directions limited by the experimental resolution, as demonstrated by the grey scale image taken at one of the sharp resonances. Similar intensity distributions are recorded for the other sharp peaks. They are, however, along the extension of the QD array slightly offset against each other. This is shown in Fig. 4, where the intensities of two peaks measured during a high-resolution line scan, marked in the inset, are plotted. The displacement of the two curves of about 190 nm, a value well within the length of the QD array, directly proves that the distinct sharp QD peaks originate from individual spatially separated dots.

Microscopic photoluminescence spectra under resonant and close to resonant excitation of the QDs are shown in the upper panel of Fig. 5. For excitation at energy (4), slightly above the emission of the dots, the QD response can be almost isolated and detected, and further narrowing of the individual sharp peaks compared to excitation at 1600 meV (Fig. 3) occurs. Resonant excitation at energy (3) results for detection at lower energies in a single PL line at position (1); i.e., recombination takes place within the lowest lying QD. Conversely, resonant excitation of this dot at energy (1) is accompanied by PL from the full series of QDs located energetically above. This anti-Stokes luminescence indicates that excitons excited in the dot which is lowest in energy can be easily transferred to the other QDs. Further insight into the intriguing interplay between the QDs can be gained by $\mu$PLE spectroscopy. For detection at energy (1) (lowest energy peak) absorption in the quantum dots and wires is probed as can be seen in the lower panel of Fig. 5. Absorption by the QDs takes place precisely at their PL energies. The fine structure in the QWR absorption coincides also with that observed in luminescence (dotted curve in the upper panel); the averaged

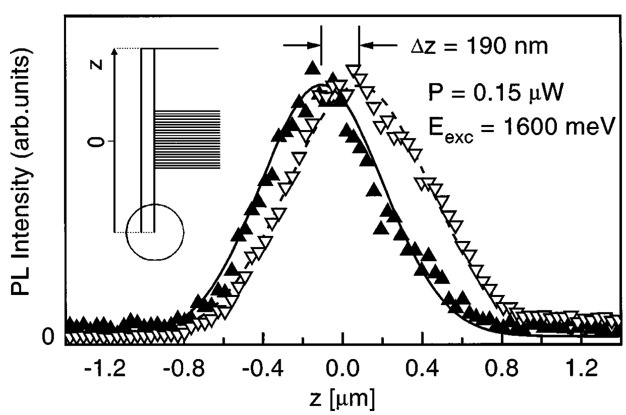


FIG. 4. High-resolution PL intensity line scans of two, spectrally well separated quantum dot peaks at 1560.7 meV (filled symbols) and 561.9 meV (hollow symbols) marked by the arrows labeled (1) and (3) in Fig. 5. The orientation of the line scan is given in the inset by the circle, indicating the detection position, which has been moved parallel to the $z$ axis along QWR$_{23}$. The solid curves are just guides to the eye.

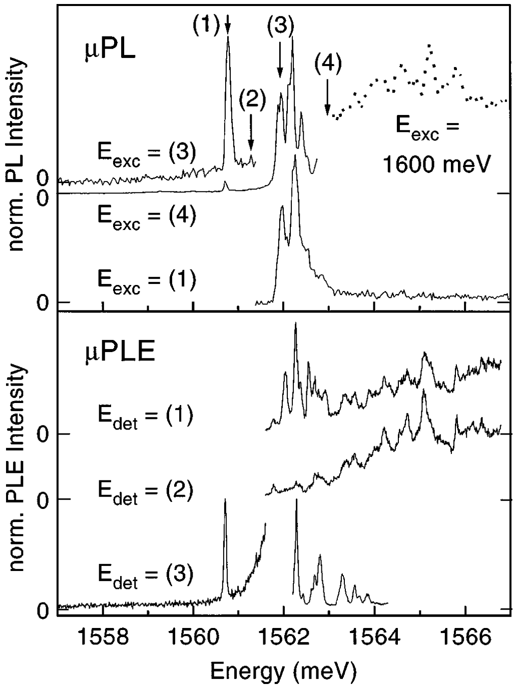


FIG. 5. $\mu$PL and $\mu$PLE spectra ($P = 0.4\ \mu$W) taken from the ($\bar{1}10$) surface at a position centered on top of the quantum dot array. The arrows mark the energetic positions used for excitation ($\mu$PL) or detection ($\mu$PLE).

peak is, however, Stokes shifted. Moving the detection energy upwards by only about 0.5 meV to the energy position (2) completely suppresses the QD features, and the $\mu$PLE spectrum is dominated by the QWR absorption. The QD absorption features reappear when the detection energy is again brought into resonance with one of the QD emission lines at energy (3). In addition, absorption by the lowest lying QD can be probed by detection at this energy. These results directly reveal the discrete nature of the 0D quantum dot states and emphasize the qualitative difference between 0D structures and systems of higher dimensionality. In order to understand the apparent strong communication between the spatially separated QDs we have performed a one-dimensional Kronig-Penney type superlattice calculation for our QD structure. As a consequence of the small barriers formed by the $QWR_{23}$ state which connects all dots, the 0D states weakly couple and a ground state bandwidth of about 1.7 meV is estimated. Consequently, the different states can be populated easily via thermal activation in the meV range.

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